

Linear-Temperature Dependence of Static Magnetic Susceptibility in LaFeAsO from Dynamical Mean-Field Theory

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In this Letter we report the LDA+DMFT (method combining Local Density Approximation with Dynamical Mean-Field Theory) results for magnetic properties of parent superconductor LaFeAsO in paramagnetic phase. Calculated uniform magnetic susceptibility shows linear dependence at intermediate temperatures in agreement with experimental data. For high temperatures (>1000 K) calculations show saturation and then susceptibility decreases with temperature. Contributions to temperature dependence of the uniform susceptibility are strongly orbitally dependent. It is related to the form of the orbitally-resolved spectral functions near the Fermi energy with strong temperature dependent narrow peaks for some of the orbitals. Our results demonstrate that linear-temperature dependence of static magnetic susceptibility in pnictide superconductors can be reproduced without invoking antiferromagnetic fluctuations.

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Introduction. LaFeAsO [1] is the first and the most investigated representative of novel iron pnictide superconductors. All known to date compounds of this class: $\text{ReFeAsO}_{1-x}\text{F}_x$ (with $\text{Re}=\text{La}, \text{Ce}, \text{Nd}, \text{Sm}, \text{Gd}$) and AFe_2As_2 ($\text{A}=\text{Ca}, \text{Sr}, \text{Ba}$) contain FeAs layers and exhibit a common phase diagram with antiferromagnetic spin-density wave (SDW) order appearing below T_N for parent compounds. The antiferromagnetism is suppressed by doping or pressure with simultaneous appearance of superconductivity. That picture resembles cuprates where superconductivity is associated with suppression of magnetism by electron or hole doping, although the undoped parent compounds for the FeAs-superconductors are not Mott-Hubbard insulators but multiband metals. These facts suggest the complex interplay of magnetism and superconductivity in iron pnictide materials.

Recently it was found that magnetic properties of these materials show anomalous behavior even in paramagnetic phase. A universal linear-temperature dependence of the static magnetic susceptibility was observed [2–6] in both parent and superconducting compounds in paramagnetic normal state above antiferromagnetic T_N or superconducting T_c transition temperatures. This non-Pauli and non-Curie-Weiss-like dependence cannot be understood within standard theories of magnetism. The authors of [2, 5] propose that this anomalous behavior appears due to the presence of a short-range antiferromagnetic fluctuations in the paramagnetic phase of iron pnictides. The antiferromagnetic fluctuations were also argued to enhance the nonanalytic (linear) term in the temperature dependence of susceptibility in a two-dimensional Fermi liquid [7], giving a possibility to provide a contribution comparable with experimental observations.

An important issue is the effect of the Coulomb interaction on the electronic and magnetic properties of iron pnictides [8–10]. It is generally accepted now that

Coulomb correlation effects in iron pnictides are not as strong as in cuprates where parent compounds are wide gap Mott insulators. Experimental spectra do not show features corresponding to Hubbard bands that are typical for strongly correlated materials. However moderate correlations lead to a significant renormalization of the electronic states near the Fermi energy with effective electronic mass value $m^*/m \approx 2$ and corresponding band narrowing of the LDA band structure is observed in ARPES [11].

In this Letter we report the first *ab-initio* computational results for uniform magnetic susceptibility of parent superconductor LaFeAsO in paramagnetic phase obtained with LDA+DMFT, which takes into account the effect of the on-site correlations. A linear increase of magnetic susceptibility is found below 1000 K with saturation and decreasing for higher temperatures. As DMFT takes into account only local correlations, the anomaly in temperature dependence of susceptibility observed in our calculations is not connected with antiferromagnetic fluctuations. It is shown that the observed temperature dependence of susceptibility can be associated with narrow temperature dependent peaks in orbitally-resolved densities of states, arising due to local correlations and located ≈ 100 meV below the Fermi energy. These peaks yield increase of the susceptibility with temperature, similar to that in metamagnetic systems. In the high-temperature regime the electrons lose their coherence and the Curie-Weiss behavior of susceptibility is restored.

Method.– The LDA+DMFT scheme proceeds in two steps: (i) construction of the effective Hamiltonian from converged LDA calculation and (ii) solution of the corresponding DMFT equations. In the present work the projection procedure onto Wannier functions [12] was used to obtain an effective 22-band Hamiltonian $\hat{H}_{\text{LDA}}(\mathbf{k})$ which incorporates five Fe d , three O p and three As p orbitals

per formula unit (two formula units per unit cell).

The DMFT self-consistent equations were solved for imaginary Matsubara frequencies. The effective impurity problem was solved by hybridization function expansion continuous-time quantum Monte-Carlo method (CT-QMC)[13] with Coulomb interaction taken in density-density form. The interaction matrix $U_{mm'}^{\sigma\sigma'}$ was parametrized by parameters U and J according to procedure described in [14]. In the present work we used $U=4$ eV and $J=1$ eV obtained via constrained LDA calculations procedure in Ref. [15]. Calculations were performed in the paramagnetic state at the inverse temperatures $\beta = 4 \div 30$ eV $^{-1}$ (from 387 to 2900 K). The energy dependence of the self-energy $\Sigma(\omega)$ on the real axis needed to calculate spectral functions was obtained by the analytical continuation via Padé approximants [16] according to procedure described in Ref. [17].

Orbitally resolved DMFT densities of states were calculated as

$$A_i(\omega) = -\frac{1}{\pi} \text{Im} \sum_{\mathbf{k}} [(\omega + \mu)\hat{I} - \hat{H}(\mathbf{k}) + \hat{\Sigma}(\omega)]_{ii}^{-1}, \quad (1)$$

where $\hat{H}(\mathbf{k})$ is the effective Hamiltonian and μ is the self-consistent chemical potential. The Hamiltonian $\hat{H}(\mathbf{k})$ is obtained by subtracting a Coulomb interaction energy E_{dc} already present in LDA (so-called double-counting correction) from d - d block of $\hat{H}_{\text{LDA}}(\mathbf{k})$. The double-counting has the form $E_{dc} = \bar{U}(n_{\text{DMFT}} - \frac{1}{2})$ where n_{DMFT} is the total number of $3d$ electrons obtained within DMFT in the absence of the external field (such approximation is justified for small fields used in uniform susceptibility calculation) and \bar{U} is the average Coulomb interaction parameter for the d shell. This choice of a double-counting term is not unique and other variants can also be used [18]. The form of E_{dc} used in the present work yields good results for transition oxide compounds [19] and pnictide materials [15, 17, 20].

The uniform magnetic susceptibility was calculated as

$$\chi(T) = \frac{\partial M(T)}{\partial E_h} = \frac{\partial [n_{\uparrow}(T) - n_{\downarrow}(T)]}{\partial E_h}, \quad (2)$$

where $M(T)$ is the field-induced magnetization and E_h is the energy correction corresponding to the applied field. For all considered temperatures the absence of polarization at zero field was checked. The derivative was computed as a ratio of the magnetization $M(T)$ and the energy E_h at small fields (where $M(T)$ is a linear function of E_h). To minimize numerical error due to Monte-Carlo procedure for each T a series of $\chi(T)$ calculations corresponding to different magnetic fields was done.

Results and discussion.— In Fig.1 we show calculated $\chi(T)$ in comparison with the experimental data of Klingeler *et al.*[2]. For temperatures below ≈ 1000 K the calculated $\chi(T)$ demonstrates roughly linear increase. To emphasize linear character of the susceptibility curve in

low-temperature region the least-square fit by a straight line to the obtained data is also shown in Fig.1. For $T > 1000$ K the susceptibility shows saturation and then decreases with temperature approaching to Curie-Weiss-like behavior at very high temperatures. To clarify the origin of the linear dependence of $\chi(T)$ in the low temperature regime it is useful to calculate orbitally resolved susceptibilities, $\chi_m = \partial M_m / \partial E_h$. Different d -orbitals of iron give diverse contributions to calculated $\chi(T)$ (see upper panel of Fig.2). The contribution of $3z^2 - r^2$ orbital possesses pronounced largest positive slope and provides the major source for linear increase of the total susceptibility below 1000 K.

It can be shown that the observed temperature dependence of $\chi_m(T)$ originates from peculiarities of orbitally resolved densities of states $A_i(\omega)$, obtained within LDA+DMFT (see Fig. 3). The calculated functions $A_i(\omega)$ are in good agreement with previously published results of Aichhorn *et al.*[21]. The correlations result in renormalization of the LDA bands with corresponding enhancement of the quasiparticle mass $m^*/m \approx 2$ and lead to appearance of peaks near the Fermi energy. The peak corresponding to $3z^2 - r^2$ orbital is especially sharp and narrow while the peaks for other orbitals are much more broad. The peaks ≈ 100 meV below the Fermi energy are absent in the LDA band structure and originate purely from the local correlations; they display strong temperature dependence, increasing in amplitude, narrowing and approaching the Fermi level with decreasing T .

These peculiarities of orbitally resolved densities of states allow to explain qualitatively the anomalous behavior of the susceptibility. The origin of increase of $\chi(T)$ with temperature is similar to that in systems with van-Hove singularities near the Fermi level[22–24], where increasing temperature yields activation of the electronic

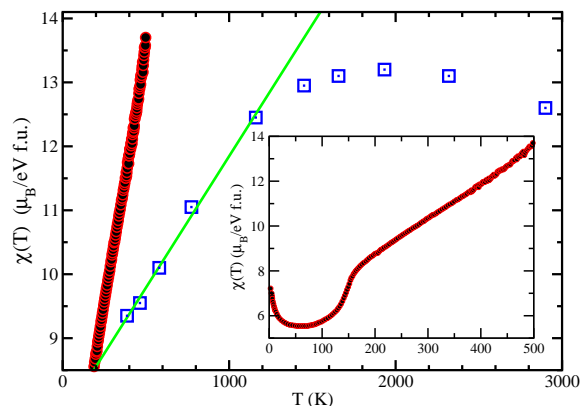


FIG. 1: (Color online) Uniform susceptibility $\chi(T)$ of LaFeAsO calculated within LDA+DMFT (squares) in comparison with experimental data of Klingeler *et al.*[2] (circles). The straight line corresponds to least-square fit of the region below 1000 K. The inset shows the full experimental susceptibility curve.

states with the energy of the peak position, which results in increase of magnetic susceptibility. When the energy $k_B T$ ($T > 1000$ K) becomes larger than distance from the peak to the Fermi level (100 meV), the linear increase of uniform susceptibility stops and the curve goes to saturation. The same mechanism applies also to the contribution of $x^2 - y^2$ orbital, which does not show peak of the spectral function, but a minimum very near the Fermi level and increase at larger energies.

To verify that the observed peculiarities of the single-particle properties are responsible for non-monotonic temperature dependence of $\chi(T)$, we have estimated the uniform susceptibility as a convolution of the DMFT interacting Green's functions neglecting vertex corrections,

$$\hat{\chi}_m^0(T) = \frac{1}{\beta} \sum_{\mathbf{k}, i\omega} \sum_{m'} \hat{G}_{mm'}(\mathbf{k}, i\omega) \hat{G}_{m'm}(\mathbf{k}, i\omega), \quad (3)$$

where $\hat{G}_{mm'}(\mathbf{k}, i\omega) = [(\omega + \mu)\hat{I} - \hat{H}(\mathbf{k}) + \hat{\Sigma}(\omega)]_{mm'}^{-1}$. Temperature dependence of the calculated susceptibilities $\hat{\chi}_m^0(T)$ is shown in the lower panel of Fig.2. The obtained curves qualitatively reproduce all features of the directly calculated susceptibilities of $x^2 - y^2$ and $3z^2 - 1$ orbitals, including their slope at low temperatures ($T < 1000$ K). Note that the off-diagonal components of

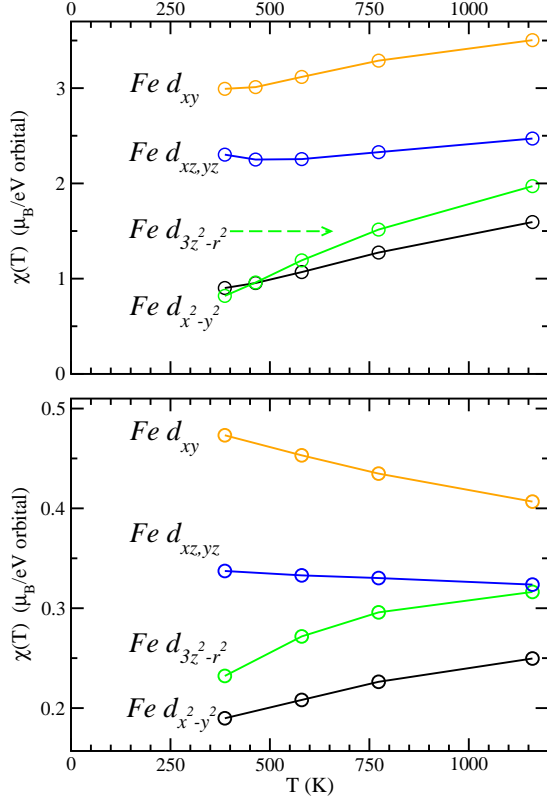


FIG. 2: (Color online) Orbitaly resolved Fe 3d susceptibilities χ_m vs temperature of LaFeAsO from the LDA+DMFT calculation obtained as derivatives of the magnetization (upper panel) and estimated via convolution of the corresponding Green's functions (lower panel).

TABLE I: Imaginary part of the self-energy (eV) at the Fermi energy E_F at different temperatures in LaFeAsO.

Temperature	387 K	580 K	1160 K
$\text{Im}\Sigma_{xy}(E_F)$	-0.142	-0.242	-0.454
$\text{Im}\Sigma_{yz,xz}(E_F)$	-0.131	-0.163	-0.306
$\text{Im}\Sigma_{3z^2-r^2}(E_F)$	-0.054	-0.092	-0.228
$\text{Im}\Sigma_{x^2-y^2}(E_F)$	-0.053	-0.101	-0.334

the Green functions also contribute partly to the linear temperature dependence of susceptibilities. For xy , xz and yz orbitals the vertex corrections appear to be more important.

The importance of the vertex corrections for some orbitals can be related to the loss of the electron coherence caused by electronic correlations, which can be traced from the electronic self-energy (Table 1). For example the imaginary part $\Sigma_{xz,yz}$ at the Fermi level is 131 meV at the lowest considered temperature and increases to 306 meV at $T=1160$ K. The loss of the electron coherence is much less pronounced for $3z^2 - r^2$ and $x^2 - y^2$ orbitals where $\text{Im}\Sigma(E_F)$ are three times smaller than the corresponding values for other orbitals at the lowest temperature.

Due to loss of coherence of electronic excitations in

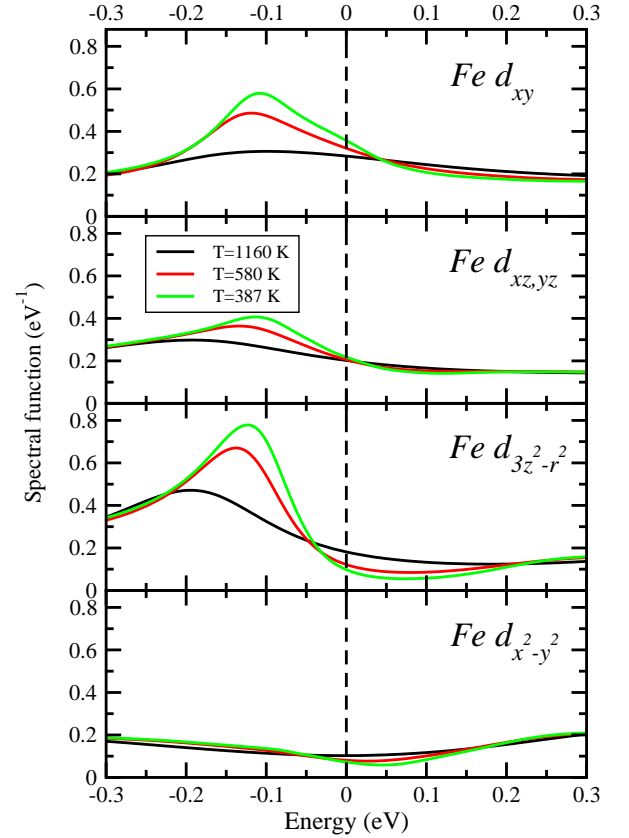


FIG. 3: (Color online) Temperature evolution of LaFeAsO Fe 3d spectral functions in vicinity of the Fermi energy (0 eV) calculated within LDA+DMFT.

part of the bands, the local susceptibility behaves very differently from the uniform susceptibility. In Fig. 4 we plot temperature dependence of the Fe- d local spin susceptibility $\chi_{\text{loc}}(T)$ and orbital contributions to its inverse value. The calculated $\chi_{\text{loc}}(T)$ decreases monotonically with temperature in agreement with the results reported in [8]. We have also calculated the mean square of the local moment $\langle m_z^2 \rangle$ as a function of temperature, which is almost temperature independent increasing approximately by only 10% of its value in the temperature region $T=387 \div 1160$ K. Analysis of the orbitally-resolved inverse susceptibilities (see inset of Fig. 4) shows that the contribution of xz and yz orbitals is almost linear in temperature, which goes along with loss of electron coherence of the corresponding states, providing a possibility of the local moment formation in these orbitals. At the same time, the electronic states originating from $3z^2 - r^2$ orbital remain more quasiparticle ($\text{Im}\Sigma_{xz,yz}(E_F) \approx 3\text{Im}\Sigma_{3z^2-r^2}(E_F)$). Such a picture is reminiscent of the proximity to the orbital-selective Mott transition, proposed recently for iron [25], iron pnictides[26] and iron oxide[27]. Quasiparticle nature of $3z^2 - r^2$ states allows to justify the above discussed scenario of the linear dependence of susceptibility due to contribution of peculiarities of the density of states in $3z^2 - r^2$ band (narrow temperature sensitive peak in spectral functions at 100 meV below the Fermi level).

Conclusion.— By employing the LDA+DMFT(CT-QMC) method we computed paramagnetic uniform magnetic susceptibility of the parent superconductor LaFeAsO. The obtained curve reproduces linear behavior at low temperatures observed in experimental data of Klingeler *et al.*[2]. We argue that the low-temperature linear increase of susceptibility in LaFeAsO comes from the presence of a sharp temperature dependent peak in the spectral function at 100 meV below the Fermi energy. Our results demonstrate that increase of the susceptibil-

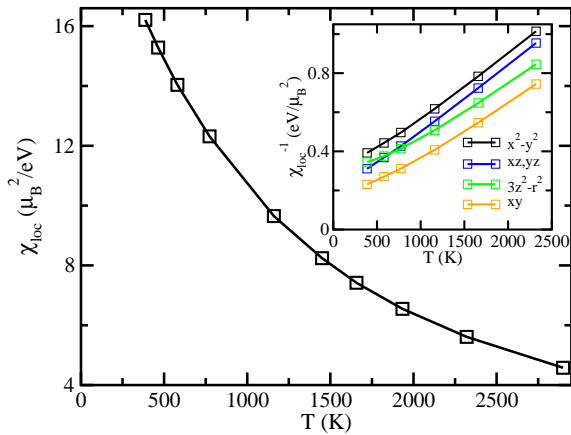


FIG. 4: The total local spin susceptibility versus temperature and temperature dependence of orbitally resolved inverse of the local spin susceptibility(inset) obtained within LDA+DMFT.

ity can be understood within single-site dynamical mean-field approach neglecting spatial magnetic fluctuations.

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